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## Study on the zeta potential effect of *Artocarpus heterophyllus* natural-based coagulant in wastewater treatment

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# Study on the zeta potential effect of *Artocarpus heterophyllus* natural-based coagulant in wastewater treatment

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**Abstract.** The application of chemical coagulants which is damaging to environment and human health has triggered to the exploration of new coagulants from natural based sources. This study emphasized on the performance of a treatment process to remove particles in the synthetic domestic wastewater by coagulation using *Artocarpus heterophyllus* (jackfruit) peel extract and evaluation of zeta potential ( $\zeta$ ). The measurement of zeta potential which is the criteria defining the electrostatic interaction between pollutants and natural coagulant agent is used in proposing mechanism of coagulation. The jar test experiment was carried out and the treated wastewater was analysed to determine reduction in turbidity, BOD, COD and TSS. The coagulant extracts were characterized in terms of surface charge. The best operating conditions were: pH 2 and dosage 60 mg/L. The corresponding removal efficiencies for turbidity, TSS, COD and BOD were 80 %, 70 %, 46 % and 20 % respectively. The measured zeta potential of the coagulant was -25.2 mV at its original pH, 6.95. On the other hand, the surface charge of synthetic wastewater was positive at pH 2 and later turned to be negative as the pH is increased up to pH 12. This best describes that jackfruit peel coagulant possessed the characteristic of anionic polyelectrolytes which involves bridging mechanism. The results obtained here suggest that coagulation using *Artocarpus heterophyllus* peel extract can be employed as an effective and low-cost preliminary technique in wastewater treatment process.

**Keywords:** wastewater, coagulation, zeta potential, *Artocarpus heterophyllus*, turbidity

## 1. Introduction

The technology of coagulation-flocculation has been practically implemented in primary treatment for contaminated wastewater. The working principle of this process is based on destabilization of colloids in wastewater and also neutralizing forces to allow agglomeration of destabilized particles [1]. In general, the widely used coagulants in treatment process are alum, ferric sulphate, polyaluminium chloride (PAC) as well as polymers comprising of synthetic and natural compounds [2-4]. The inorganic and synthetic coagulants possess several drawbacks including large amount of sludge production, requirement of pH adjustments and high concentration of aluminium in treated water which is associated to harmful effects on human health [4-6]. In order to overcome these disadvantages, application of



natural based and environmental friendly coagulants originating from plants such as *Moringa oleifera*, chitosan, starches, and tannin has been explored as an alternative solution [7-8]. Natural coagulants are safe to the environment and less sludge is produced [9]. This type of coagulant can be easily manifested as point-of-use technologies in population which lack in development as this utilizes natural resources of locally-available and low cost [10-11].

Various factors influences the effectiveness of coagulation-flocculation process. The surface charge of suspended particles with dissolved metals is one of the important parameters under consideration as it is a factor hindering the solid and metals separation. Zeta potential ( $\zeta$ ) is defined as interfacial parameter linked condition of dissolved metals at low solubility when the point of isoelectric is attained. However, there is no direct measurement of zeta potential. The zeta potential can be determined by measuring the mobility of the particles using techniques such as electrophoretic mobility as well as using theoretical models. Electrophoresis can be described as the motion of particles dispersion relative to a liquid under the influence of electric field which is spatially uniform. It was reported by Meraz et al. (2016), greater removal of organic matter was obtained at certain value of zeta potential and deduced that the interactions existing between the natural coagulants (biopolyelectrolytes) and wastewater were due to Van der Waals interactions and forces of electrostatic [12]. The zeta potential ( $\zeta$  [ = ] mV) for slurry particles is commonly used as a benchmark to evaluate the stability of particles' in terms of colloidal. Zeta potential can be well interpreted as electric potential at the slipping plane with conjunction to a point in the bulk medium. Nevertheless, zeta potential is neither equivalent to the surface potential nor Stern potential as they are defined in terms of different locations around the particle. Those ions enclosed within the slipping plane and Stern layer are mainly transported along with the particle. Thus, the effective charge on the particles is often reviewed as the zeta potential. The electric double-layer are referred to the disintegration of these two layers and approaching the potential of the bulk solution. Particles with like charges are prone to a lower propensity for agglomeration as a result of increase in absolute zeta potential. In summary, slurries or composition of mixture with particle zeta potentials of  $\zeta > |30|$  are regarded as stable colloids [13].

Current study was performed to evaluate the performance of *Artocarpus heterophyllus* as natural coagulant in wastewater treatment. Initial pH of wastewater, coagulant dosage and zeta potential were the variables analyzed in this study. The performance of this natural coagulant was determined by measuring the turbidity, total suspended solids (TSS), biochemical oxygen demand (BOD) and chemical oxygen demand (COD) after the treatment. It was anticipated that the novel coagulant extracted from *Artocarpus heterophyllus* peel applied in this study would contribute to a beneficial approach for other challenging systems in the field of environmental.

## 2. Experimental

### 2.1. Materials

The main raw material which is *Artocarpus heterophyllus* peels were collected from fruit stall located in Pahang, Malaysia. The chemicals used were sodium hydroxide (NaOH) and analytical-grade hydrochloric acid (HCl) which were purchased from Fisher Scientific Malaysia. The cat food used in preparation of synthetic wastewater was a commercial brand known as Whiskas® (Ocean Fish Flavor) obtained from hypermarket.

### 2.2. Synthetic Wastewater Preparation

The preparation is carried out by grinding cat food pellets into powder form. Then, 10 grams of ground cat food were mixed into 1 litre of tap water. This synthetic wastewater simulates the domestic wastewater of medium strength. [14]. Synthetic wastewater was prepared freshly each time jar tests were carried out. The nutritional constituents of this cat food includes 30 % crude protein, 12 % moisture, 10 % crude fat and 5 % crude fiber. The prepared synthetic sewage were classified based on different parameters as shown in Table 1. The methods used in measurement of BOD, COD,  $\text{NH}_3\text{-N}$ ,  $\text{NO}_3\text{-N}$  and phosphorus were based on APHA 5210B, APHA 5220D, APHA 4500 ( $\text{NH}_3\text{-N}$ ), APHA 4500 ( $\text{NO}_3$ ) and

APHA 3010 respectively. TSS test was performed with vacuum filtration method. The purpose of using synthetic sewage in this study was to retain and control the wastewater properties. This allows the initial condition of wastewater to be controlled unlike real wastewater that always change depending on the type of waste that is processed. Turbidity of wastewater was analyzed with LaMotte portable turbidimeter which is measured in nephelometric turbidity unit (NTU).

**Table 1.** Characteristics of synthetic wastewater

Parameter	Synthetic wastewater (mg/L)	Real domestic wastewater (mg/L) [27]
BOD	190	300
COD	430	1500
TSS	210	216
NO <sub>3</sub> - N	27	40
NH <sub>3</sub> - N	15	25
Phosphorus	42	7

### 2.3. Natural Coagulant Preparation

The collected peels of *Artocarpus heterophyllus* were cleaned thoroughly using distilled water to eliminate impurities. The peels were cut into smaller pieces approximately within the size of 4 cm to 5 cm. The peels were then dried for 48 hours at 60 °C in the oven (Memmert Model 30, Germany) to remove the moisture content. The dried peels were ground finely and passed through a sieve of 0.5 mm to ensure uniformity in particles size. The dried raw materials of mass 0.5 g was soaked in 100 ml of distilled water. This mixture was stirred for 1 hour at a speed of 120 rpm using magnetic stirrer. This preparation was performed at room temperature of  $25 \pm 2$  °C. The suspension was filtered with muslin cloth and the liquid form of the filtrate was used in the jar test experiments [15-17].

### 2.4. Zeta Potential Analysis

Zeta potential of the extracted coagulant and wastewater were measured using zeta analyzer (Malvern Zetasizer Nano ZS Series, UK) which is a light scattering equipment to identify the surface charges of the coagulant extract. This analysis was conducted at a constant temperature of 25 °C.

### 2.5. Jar-Test Experiment

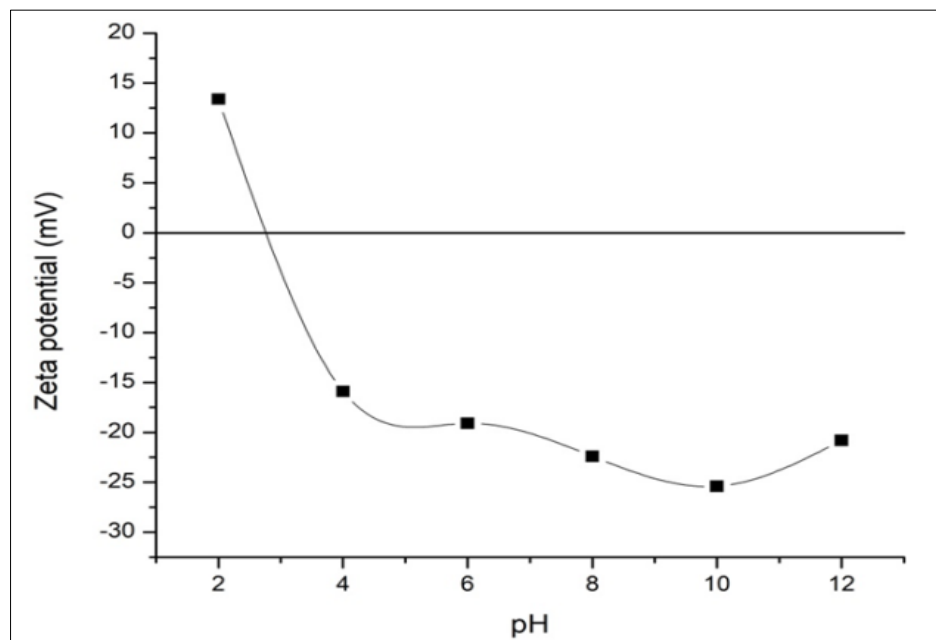
The jar floc test method were used for coagulation test experiments by utilizing six-paddle rotor jar test equipment (JLT6 Velp Scientifica, Italy). The beakers were filled with 500 ml of prepared wastewater and respective amount of natural coagulant were added to run for the jar test. The jar test was carried out at room temperature of 25 °C. The jar-test was performed with different pH of wastewater within range of pH 2 – pH 12. The pH of wastewater is modified as required by adding 1.0 M NaOH and 1.0 M HCl. All of these analyses were performed in triplicate. In terms of dosage of the coagulant, it was fixed at 100 mg/L for this experiment. The initial turbidity was recorded for every sample. The suspension was stirred with a condition of rapid mixing for 4 minutes at 100 rpm and succeeded by slow mixing at 40 rpm for 25 minutes. This is to allow flocs particles to uniformly suspend. The mixture was then left for 1 hour for the process of settling. The supernatant was withdrawn using pipette from 2 cm below the liquid level to proceed with the water quality analysis. The turbidity, TSS, BOD and COD reduction was calculated for each sample. The same steps were repeated in order to analyse the effect of dosage within range of 20 mg/L to 120 mg/L.

### 3. Results and discussion

#### 3.1. Evaluation of zeta potential measurement

The zeta potential measurements is greatly influenced by pH especially in aqueous dispersions that correlates with the coagulation process of wastewater. Fig. 1 indicates the zeta potential of wastewater by varying the pH. The measured zeta potential of the coagulant was -25.2 mV at its original pH, 6.95. Zeta potential varies with pH where the magnitude becomes more positive at acidic condition and more negative at alkaline condition [18]. The surface charge of the synthetic wastewater is positive at pH 2 and later turned to be negative as the pH is increased up to pH 12. The addition of alkali which was sodium hydroxide to the wastewater caused the particles to be negatively charged. Contrarily, when acid is added, negative charge is neutralized to the point where the zeta potential achieved is zero. By further adding the acid, a build-up of positive charges in the particles of wastewater took place.

The magnitude of zeta potential indicates whether colloidal-size particle in suspension will repel one another and remain in suspension or agglomerate and eventually settle. Particles in suspension that possess a large positive or negative value of zeta potential will have the tendency to repel each other and will not agglomerate. On the other side, particles with smaller value of zeta potential will flocculate and form bigger colloids. The conditions where zeta potential deviates from zero to optimum coagulation indicates other coagulation mechanisms contributing to the coagulation process instead of charge neutralization [19]. Thus, it can be described that jackfruit peel coagulant possesses the characteristic of anionic polyelectrolytes where the primary mechanism involved in the aggregation of the constituents was bridging. This can be correlated with Zhang et al. (2010) revelation, where addition of anionic flocculants (TJ-FI) that is from a mixed activated sludge produced by *Proteus mirabilis* induced negative zeta potential of kaolin clay suspension [20]. This resulted in static repulsive forces to be increased among the particles suggesting that bridging mechanism.



**Figure 1.** Variations of zeta potential with respect to pH of synthetic wastewater

#### 3.2. Effect of initial pH of wastewater

As for this experiment, the pH of the synthetic domestic wastewater as well as the dosage of coagulant were altered as required before the coagulation test was performed with *Artocarpus heterophyllus* peel extract based coagulant. The pH of wastewater mainly affects the surface charge of coagulants and also

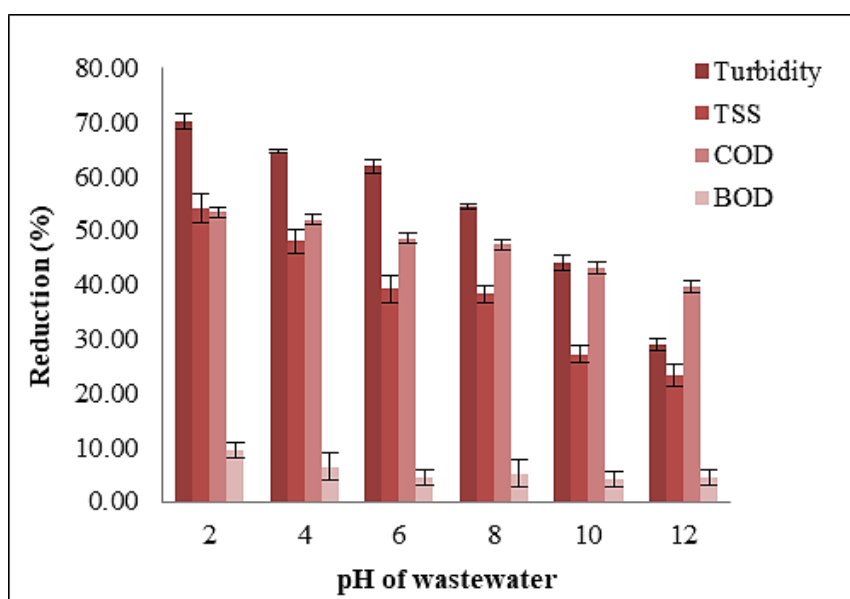
the stabilisation of suspension. The results obtained from the coagulation test by using *Artocarpus heterophyllus* peel extract for varied pH of wastewater is as shown in Fig. 2.

As can be seen, the coagulation performance of *Artocarpus heterophyllus* peel in reducing turbidity favoured the acidic condition. It was observed that the *Artocarpus heterophyllus* peel extract worked the best as a coagulant at pH 2. All the wastewater parameters that were tested including turbidity, TSS, COD and BOD showed significant reduction at this particular pH. Based on this graph, it can be observed that the effectiveness of the coagulant decreased as the wastewater turns alkaline. The percentage of reduction obtained were  $70 \pm 1 \%$ ,  $55 \pm 2 \%$ ,  $53 \pm 2 \%$  and  $10 \pm 2 \%$  respectively. The trend from Figure 2 suggested that the increase in pH, decreased the percentage of reduction of tested parameters. It also can be deduced that BOD reduction is not apparent by using this natural coagulant as this parameter relies on biological treatment.

Previous studies have been conducted by applying different natural coagulant on wastewater treatment with varied pH. Shamsnejati et al. (2015) have reported on usage of seed of *Ocimum basilicum* for textile wastewater treatment and found that maximum COD reduction was at pH 6.5 which is in acidic condition as well [21]. Besides, okra mucilage has also been tested on textile wastewater and was discovered that significant reduction obtained at pH 5. Reduction of COD and BOD obtained were 48 % and 92 % respectively at acidic condition [22]. In another study, using banana pith, it was discovered that this natural coagulant works best at pH 4 where turbidity and COD reduction 98 % and 54 % respectively [23]. Most of the natural coagulants from literature studies revealed that they are effective in acidic condition. These findings coincided to the current study that proved *Artocarpus heterophyllus* peel extract coagulant also worked effectively at pH within the range of acidic condition.

The effectiveness of the coagulant is evaluated based on the clarity of wastewater and colloids formed after the treatment. As for this study, the water appeared to be the clearest at pH 2 and more colloids settlement were observed that improved the clarity of the wastewater. The effectiveness starts declining gradually when the pH is increased towards alkaline condition. The water turns cloudier with percentage of turbidity reduction in the range of  $67 \pm 1 \%$  to  $29 \pm 1 \%$  and less flocs were found after the settling process due to pH incremental. The lowest turbidity reduction was found at pH 12 when the wastewater condition is most alkaline. The turbidity reduction was only  $30 \pm 1 \%$  which indicates the least effective pH for *Artocarpus heterophyllus* peel to perform as coagulant. The BOD, COD and TSS removal declined as the pH is increased. At pH 12, the percentage reduction of COD, BOD and TSS were observed to be  $23 \pm 2 \%$ ,  $40 \pm 3 \%$  and  $4 \pm 2 \%$  respectively. The coagulative behaviour of *Artocarpus heterophyllus* peel decreases when the alkalinity of the wastewater increases as within the range of pH 8 to pH 12. Thus, it clearly shows that *Artocarpus heterophyllus* peel works well in acidic condition which supported with characterization analysis.

Based on zeta potential study, the zeta potential measured was -25.2 mV at pH of 6.95 which indicates that the surface charge present in the coagulant is negative. As reported by Lestari et al. (2010), the acidic condition of wastewater promotes the positive charges on molecules that may influence and enhance the performance of the molecules to function well as coagulant [24]. Based on Fig. 1, it can be clearly observed that the colloids in the wastewater possess positive surface charge which promotes agglomeration between colloids. Acidic condition of wastewater is highly present with hydrogen ions which also increases the positive charge of the wastewater. Thus, this phenomenon significantly affects the turbidity reduction of the synthetic wastewater. In acidic pH condition, negatively charged groups of the coagulant are protonated and experience neutralization process as well. Furthermore, the particles surface charge undergoes neutralization and destabilization took place. This mechanism consequently contributes to coagulation process [21].



**Figure 2.** The percentage of reduction against varied pH of wastewater using *Artocarpus heterophyllus* peel extract.

### 3.3. Effect of coagulant dosage

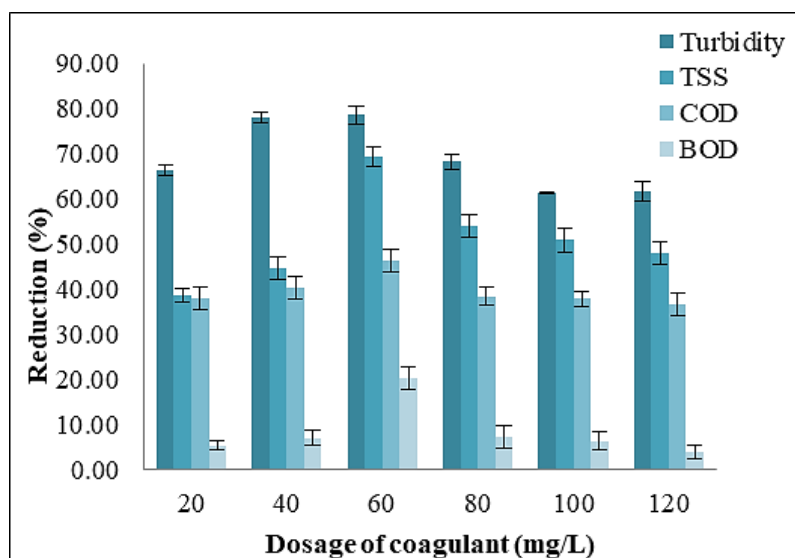
Coagulant dosage another important factor that has been evaluated in order to determine the optimum condition for coagulation process. Intrinsically, insufficient dosage or overdosing of coagulant would contribute to poor performance and efficiency in the process of coagulation. Hence, it is significant to analyse the optimum dosage as the dosing cost of extraction process can be minimized, reduction of sludge formation and obtain optimum performance in treatment. This experiment was carried out by maintaining the pH of wastewater at 2 as obtained in previous study. Fig. 3 displayed the trend of turbidity reduction at different dosage of *Artocarpus heterophyllus* peel.

The optimal dosage of the coagulants were examined by varying the coagulant dosage at optimum pH. The coagulant dosage used was ranging from 20 mg/L to 120 mg/L. Figure 3 revealed that the value increases steadily when the dosage was increased up to 60 mg/L. Highest removal achieved for turbidity, TSS, COD and BOD were  $80 \pm 2\%$ ,  $70 \pm 2\%$ ,  $46 \pm 2\%$  and  $20 \pm 2\%$  respectively. Condition of water appeared to be the clearest and bigger flocs were observed in the sample test. The coagulant effectiveness started declining gradually when the dosage was increased from 80 mg/L up to 120 mg/L. The water turns cloudier and less flocs were found after the settling process. The reduction percentage of TSS, turbidity, COD and BOD also declined justifying that overdosing of coagulant occurred at this range. The optimum dosage that the *Artocarpus heterophyllus* peel can be most effective was at 60 mg/L and further increased in dosage did not improve the removal percentage.

From this study, it can be interpreted that the dosage of coagulant highly influences the process of coagulation. This is because increase in the dosage of coagulant, increases the effectiveness in reducing turbidity of the wastewater but limited to its optimum coagulant dosage. The optimum dosage of the coagulant resulted in highest turbidity removal. It is perceived that the *Artocarpus heterophyllus* peel extract works the best at the dosage of 60 mg/L. Under dosing of coagulant lowers the effectiveness in reducing turbidity and other pollutants in the wastewater. It was observed that the wastewater appear cloudier with less colloids formation at coagulant dosage of 20 mg/L. The particles were still found in the wastewater which did not settle to the bottom of the beaker. While overdosing of coagulant may results in the re-dispersion of impurities due to the charge reversal. This will cause the breakage of colloids that increases the turbidity of wastewater.

In this result, it can be deduced that concentration of coagulant above the optimum may hinder the reaction between the coagulant and particles in the wastewater. This reduction can be inferred that

overdosing contributes to destabilization of colloidal particles and charge reversal. Hydrolyzation of coagulant in wastewater produces cationic species which can be absorbed by particle with negative charges and neutralize their charge. Hydrolysis is a reaction between organic chemical and water to form new substances. This commonly involves the chemical bonds cleavage with addition of water. In addition, hydrolysis can also be the reverse reaction of a condensation process where two molecules join together into a larger one and expel a water molecule [25]. Thus, the mechanism of particles destabilization allows flocculation process to take place and coagulant overdosing may interfere this process [26].



**Figure 3.** The percentage of reduction against varied dosage of coagulant using *Artocarpus heterophyllus* peel extract.

#### 4. Conclusion

The results obtained from the present study show the feasibility of locally available *Artocarpus heterophyllus* peel as natural coagulant on the removal of turbidity, BOD, COD and TSS in synthetic wastewater. Highest removal achieved for turbidity, TSS, COD and BOD were  $80 \pm 2\%$ ,  $70 \pm 2\%$ ,  $46 \pm 2\%$  and  $20 \pm 2\%$  respectively. The zeta potential measurements implied the correlation of pH and colloidal formation in eliminating contaminants in wastewater. *Artocarpus heterophyllus* peel coagulant can be used as primary treatment of the wastewater. This coagulant is believed to be an environmental friendly alternative to existing chemical coagulants that are currently being practiced in wastewater treatment.

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